Uranium Uptake Study,

Nambe, New Mexico: Source Document

Los Alamos
N A T I O N A L   L A B O R A T O R Y

Uranium Uptake Study,
Nambe, New Mexico: Source Document

A. C. Hayes
P. R. Fresquez
W. F. Whicker[1]

[1]*Department of Radiological Health Sciences, Colorado State University, Fort Collins, CO 80523*

Los Alamos

N A T I O N A L   L A B O R A T O R Y

Los Alamos, New Mexico 87545

Contents

List of Tables

URANIUM UPTAKE STUDY, NAMBE, NEW MEXICO: SOURCE DOCUMENT

A.C. Hayes, P.R. Fresquez, and W.F. Whicker

ABSTRACT

Over 50% of the wells in the Nambe region of northern New Mexico exceed the U.S. Environmental Protection Agency's recommended drinking water standard of 20 µg L^{-1} for ^{238}U. Uranium uptake was estimated in tomato (*Lycopersicon esculentum*), squash (*Cucurbita pepo*), lettuce (*Lactuca scarriola*), and radish (*Raphanus sativus*) irrigated with Nambe well water containing <1, 150, 500, and 1200 µg U L^{-1}. Plant uptake and human dose and toxicity associated with ingestion of water and produce and inhalation of irrigated soil related to gardening activities were evaluated. Uranium concentration in plants increased linearly with increasing U concentration in irrigation water, particularly in lettuce and radish. The estimated total committed effective dose equivalent for 70 years of maximum continuous exposure, via the three pathways to well water containing 1200 µg U L^{-1}, was 0.17 mSv (17 mrem) with a corresponding kidney concentration of 0.8 µg U g^{-1} kidney.

1.0 INTRODUCTION

1.1 STATEMENT OF PROBLEM

During the summers of 1975, 1976, and 1978, Los Alamos Scientific Laboratory, now known as Los Alamos National Laboratory (LANL), conducted hydrogeochemical sampling for the National Uranium (U) Resource Evaluation project (Maassen and Bolivar, 1979). Water and soil samples were collected in the Albuquerque quadrangle. In addition to including portions of Rio Arriba, Santa Fe, Torrance, Bernalillo, Valencia, McKinley, and Sandoval Counties, this region encompasses the entire County of Los Alamos.

All 408 water samples, collected from streams, wells, and springs, were analyzed for numerous elements including U, which ranged from below detection (0.02 µg U L^{-1} [ppb]) up to 194.06 µg U L^{-1}. Water samples containing the highest U concentrations were collected near Pojoaque, in the Nambe region of northern New Mexico, including a stream sample containing 194.06 µg U L^{-1} and a well water sample containing 94.84 µg U L^{-1}.

In 1995, the Ground Water Protection and Remediation Bureau of the New Mexico

Environment Department (NMED) sampled 72 private wells in the Nambe region (McQuillan and Montes, 1998). Thirty-seven (52%) of these wells contained U concentrations exceeding the U.S. Environmental Protection Agency's (USEPA) proposed limit of 20 µg U L^{-1}. U concentrations were found to range from below detection limits up to 920 µg U L^{-1}.

Currently, numerous wells in the Nambe area are used as primary sources of drinking water and irrigation for home garden production. Daily utilization of these wells, coupled with the elevated levels of U, has raised concerns of potential radiological and toxicological risks to human consumers.

1.2 PURPOSE OF STUDY

This study evaluated site-specific radioecological issues relating to exposure via the well water pathway in the Nambe area. The well water pathway is defined to include ingestion of well water, ingestion of vegetables irrigated with well water, and inhalation of soils irrigated with well water. The study was designed to address two primary questions:

1)   Do vegetable crops irrigated with Nambe well water accumulate U above background concentrations?

2)   What are the radiological and toxicological risks to human consumers exposed to the well water pathway in the Nambe area?

The question relating to plant accumulation was addressed for two reasons. First, there is very little published data on plant uptake of soluble U. A risk assessment utilizing concentration ratios (CRs) representing plant uptake of insoluble U was considered unrealistic. Secondly, published data indicate that CRs reflecting U uptake by crop species are highly variable and site specific (Sheppard and Evenden, 1988), as well as dependant upon plant species (Mahon, 1982). An accurate risk assessment would require CRs representing crop species commonly grown in the Nambe region.

Addressing the radiological and toxicological risks was necessary for two reasons. First, the USEPA has proposed a limit of 20 µg U L^{-1} in drinking water based on kidney toxicity (Ambika Bathija, personal communication 11/98). Performing a risk assessment based on the well water pathway, as defined above, will allow evaluation of the proposed limit.

A risk assessment was also needed to address concerns expressed by homeowners of the affected wells. The risk assessment will provide quantitative information that can be used by State and Federal regulators, as well as the public, and will contribute to

scientific knowledge concerning U uptake and associated risks from exposure to elevated levels in drinking water.

Data were obtained by establishing an experimental garden to approximate normal growing conditions for the Nambe region of northern New Mexico. Samples of produce from the experimental garden, collected at the end of the growing season, were analyzed for numerous elements including U. The resulting data were utilized to assess root uptake as a function of U concentration in irrigation water and the associated risks to human consumers.

2. LITERATURE REVIEW

2.1 GEOCHEMISTRY

U is a primordial radionuclide created during the formation of the earth, and, with a 4.5-billion-year half-life, it is ubiquitous throughout the environment. It is present in all rocks and soil including granite, metamorphic rocks, lignites, monazite sand, and phosphate deposits (Hess et al., 1985) with an average concentration in the continental crust of 2.8 ppm (Eisenbud and Gesell, 1997). U is the heaviest naturally occurring element (Cothern and Lappenbusch, 1983) and is classified as an actinide, which are elements occurring between actinium (atomic number 89) and rutherfordium (atomic number 104) (Kotz

and Purcell, 1991). Natural U is a composite of three isotopes including ^{238}U (99.28%), ^{235}U (0.71%), and ^{234}U (0.0058%) (Eisenbud and Gesell, 1997). The ^{238}U isotope, the parent of ^{234}U and the "uranium series" (NCRP 94, 1987), exists in radioactive equilibrium with ^{234}U, while ^{235}U is the parent of the actinium series (Eisenbud and Gesell, 1997). All three U isotopes decay in series and eventually result in a stable lead isotope (Cowart and Burnett, 1994).

U occurs in five valence states: +2, +3, +4, +5, and +6 (Cothern and Lappenbusch, 1983). The uranous (+4) and uranyl (+6) oxidation states are found in the environment (De Vivo et al., 1984). Uranous complexes are found in reduced environments while uranyl complexes are typically found in oxidized environments (De Vivo et al., 1984).

2.1.1 The Uranous Ion (U IV)

De Vivo et al. (1984) provide an excellent description of uranous complex geochemistry. Solubility of uranous complexes is primarily dependent on temperature and pH. Environmental transport of uranous compounds becomes important above 150°C where the predominant species are formed. Given the appropriate ligand concentrations, these species include uranous fluoride, phosphate, sulfate, and especially

hydroxide compounds. In terms of pH, uranous complexes are insoluble between pH 4.5 and pH 7. Titayeva (1994) confirms this by stating that the uranous ion is a weak base and is present in only very acidic solutions. Only two uranous complexes, UCl_4 and $U(SO)_4$, are both soluble and stable (Titayeva, 1994).

2.1.2 The Uranyl Ion (U VI)

In contrast to the relatively insoluble and unstable uranous complexes, uranyl complexes are soluble over a wide range of conditions (De Vivo et al., 1984). Uranyl ions easily form complexes and hydrolyze instantly in water to form the complex cation UO_2 (Titayeva, 1994). NCRP 94 (1987) identifies numerous factors affecting the solubility of uranyl complexes. These factors include pH, temperature, redox potential (closely tied to the dissolved oxygen content), concentration of dissolved solids, and flow rate, which can be discounted discussing ground water (NCRP 94, 1987). In $25^{o}C$ water, uranyl fluorides are formed at a pH <4, uranyl phosphates are formed at a pH 4–7.5, and uranyl di- and tri-carbonate complexes are formed at a pH >7.5 (De Vivo et al., 1984). The broad range of U concentrations in ground water is primarily due to the change in solubility that occurs at an Eh of approximately -0.1 to -0.2 (Hostetler and Garrels, 1962) and pH of approximately 6 (Dement'yev and Syromyatnikov, 1968).

The most important complexes formed by the uranyl ion are the carbonate, sulfate, fluoride, phosphate, and hydroxyl complexes (Titayeva, 1994). Water having an excess of CO_3^{2} results in the formation of the tri-carbonate complex $[UO_2(CO_3)_3]^{4-}$ which yields the di-carbonate complex $[UO_2(CO_3)_2(H_2O)_2]^{2-}$ upon dissolution (Titayeva, 1994). In Nambe, the predominant uranyl species in ground water is sodium-bicarbonate-sulfate (McQuillan and Montes, 1998).

2.2 URANIUM IN GROUND WATER

Physical and chemical availability of U in the environment make U a common constituent in natural waters including seawater, freshwater, and surface and ground water. The average concentration of U^{238} in seawater is approximately 0.12 μg U L^{-1} and roughly 0.5 μg U L^{-1} in freshwaters such as streams and rivers (Eisler, 1994). Most drinking water supplies contain natural uranium levels below 3 μg U L^{-1} (ATSDR, 1990), although elevated levels of natural U have been discovered in both surface and ground water (Hess et al., 1985). A number of states have found average drinking water levels exceeding 2 pCi L^{-1}, the equivalent of roughly 6 μg U L^{-1} (Irwin et al., 1997).

These states include South Dakota, Nevada, New Mexico, California, Wyoming, Texas, Arizona, and Oklahoma. A survey of 28,239 surface and ground water supplies from which drinking water had been obtained was conducted. Results found 2228 of the water supplies contained 10 pCi U L^{-1} (30 μg U L^{-1}) or more while 979 contained 20 pCi L^{-1} (60 μg U L^{-1}) or more. Small towns serving less than a few thousand persons generally use these water sources (Cothern and Lappenbusch, 1983). The predominant U isotopes in naturally occurring water are ^{238}U and its daughter, ^{234}U.

2.2.1 Drinking Water Regulations

Current regulatory guidelines, relating to radioactivity in drinking water, have been in effect for over 20 years. In 1976, the National Interim Primary Drinking Water Regulations were announced but did not establish recommended maximum contaminant levels for many radionuclides in drinking water (Aieta et al., 1987). The USEPA issued an Advance Notice of Proposed Rulemaking for radionuclides in drinking water in 1983 to establish maximum concentration level goals (MCLGs) and maximum concentration levels (MCLs) for ^{226}Ra, ^{228}Ra, natural U, radon, gross alpha, gross beta, and photon emitters (Aieta et al., 1987). The announcement of the new

MCLGs and MCLs was anticipated to be sometime in 1989 with compliance becoming mandatory within 18 months of the announcement (Aieta et al., 1987). In 1986, amendments to the Safe Drinking Water Act required that a standard be set for U (USEPA, 1996a). In 1991, the USEPA announced proposed guidelines for radionuclides, including U, and by Court order, the proposed standards must be finalized or the existing standards must be ratified by November 2000 (USEPA, 1996a).

The 1991 proposed MCLG for U in drinking water is 0 pCi L^{-1}, based upon its carcinogenic potential classification (Group A), while the proposed MCL is 20 μg U L^{-1} (USEPA, 1996b). In contrast to these limits, ICRP 64 recommended a limit of 1800 μg U L^{-1}, based upon chemical toxicity, while ICRP 79 recommended a limit of 4770 μg U L^{-1} based on bone cancer risk (Wrenn et al., 1985). The Department of Energy (DOE) has established a limit of 800 μg U L^{-1} for natural U based on the Department's public dose limit guideline for the general public (ESP, 1996). Surface and ground water drinking sources may contain maximum concentrations between 582 pCi L^{-1} and 653 pCi L^{-1} (403 μg U L^{-1} and 452 μg U L^{-1}, respectively)(ATSDR, 1990). Wrenn et al. (1985) point out the use of a safety factor

between 50 and 150 when establishing regulatory guidelines for natural U. Applying a safety factor of 100 to the ICRP 64 recommended limit of 1800 μg U L^{-1} results in a limit of 18 μg U L^{-1}, which is roughly equivalent to the 20 μg U L^{-1} recommended by the USEPA.

2.3 PLANT UPTAKE

Plant uptake is directly dependent on soil composition and plant metabolism and indirectly dependent on numerous environmental factors including physiological conditions and the influence of root systems on ion uptake from the soil (Osburn, 1965). That U serves no metabolic function in plants is generally accepted (Eisenbud and Gesell, 1997). However, at low substrate concentrations, U may be utilized by higher plants as a micronutrient (Sheppard and Sheppard, 1985; Cannon, 1952). U follows the translocation pathway of phosphorous (Sheppard and Evenden, 1988) for which it is considered an analogue (Mengel and Kirkby, 1979). U has been found to compete with Ca^{2+}, Ba^{2+}, and Ra^{2+}, which can complicate the uptake process (Linsalata, 1994). Mortvedt (1994) states that calcium content in plants is genetically controlled and appears to be a passive process with calcium being preferentially translocated to the plant's shoot apex with

very little being transported to the fruit and storage organs. Supporting evidence of the competition between calcium/uranium is indicated by Drobkov (1951), who found the highest U concentration (2800 μg U Kg^{-1}) in grape seeds, confirmation of U accumulation in the fastest growing plant portions, and the competition between uranium and calcium. Sheppard and Evenden (1988) and Dunn (1986) found U concentrations tend to rank in the following order: twigs > leaves > roots > trunk. A process limiting U uptake is adsorption on cell wall materials, which can result in higher concentrations in lower parts of the plant (Sheppard and Evenden, 1988). This restricts U, Th, and Pb uptake and can result in root surfaces having the highest concentrations (Sheppard and Evenden, 1988).

2.3.1 Concentration Ratios

The transfer of radionuclides between ecological and biological compartments is commonly described in terms of CR. CRs describe the amount of a radionuclide entering a biological compartment, such as vegetation, from an ecological compartment, such as soil. The following two assumptions must be made in order for the CR concept to be valid. First, that the element of interest is in equilibrium in each compartment, and, second, that the CR is constant under similar

conditions regardless of substrate concentration (Sheppard and Sheppard, 1985). The CR concept is both convenient and useful but caution must be exercised due to inherent errors in its estimation. Numerous authors have dealt with the complex issues surrounding CRs (Sheppard and Sheppard, 1985; Simon and Ibrahim, 1987; Sheppard and Evenden, 1988; McGee et al., 1996).

Comparing CRs obtained from research is both a monumental and difficult task. This is due to complex biological interactions involved in plant uptake based upon plant species, site-specific variables, and the unlimited experimental methodologies utilized in these studies. Sheppard and Evenden (1988) attribute wide ranging uranium CRs, which vary from three to five orders of magnitude, to the dynamic physical, chemical, and biological interactions in the plant/soil system. For example, CRs have been estimated based on a variety of plants including native and crop species that have been grown in-situ or ex-situ with different soil types and volume using different U concentrations in many different forms. This illustrates why such wide ranges of CR values are observed instead of the theoretically constant CR and why CR values are site specific.

Although site-specific CRs are often not available, both the U.S. Nuclear Regulatory Commission (Kennedy and Strenge, 1992) and the International Atomic Energy Agency (IAEA) (1994) have published CR recommended guidelines for radionuclides (Table 1). A comparison of the recommended values shows inconsistencies in both the vegetation categorizations and in CR values. IAEA Technical Report No. 364 (1994) recommends CRs, for U uptake, between 10^{-2} to 10^{-3} with an uncertainty factor of 10. This results in an overall range of 10^{-1} through 10^{-4}.

Table 1. Recommended Concentration Ratios

VEGETATION TYPE	NUREG/CR-5512	IAEA Tech. Report No. 364
Leafy Vegetables	1.7×10^{-2}	---
Root Vegetables	1.4×10^{-2}	---
Mixed Green Vegetables	---	8.3×10^{-3}
Mixed Roots (Roots)	---	1.4×10^{-2}
Potato (Tuber)	---	1.1×10^{-2}

Two publications were found (Lakshmanan and Venkateswarlu, 1988, and Morishima et al., 1977) that dealt with U uptake in plants from water containing elevated concentrations of U. Both of these indicate that CR values representing U uptake from water exceed 1.0 while CR values representing U uptake from soil generally fall within the recommended range of 10^{-2} to 10^{-3} (IAEA, 1994).

Lakshmanan and Venkateswarlu (1988) studied uptake from U amended soils and well water containing U in vegetables and rice. Results from this study found that CRs for the U amended soil ranged from 10^{-3} to 10^{-4} while CRs for the water with elevated U concentrations ranged from 10^{-2} to 10^{1}. The estimated CRs in this study were based on the fresh weight of vegetation. Morishima et al. (1977) found that CRs reflecting U uptake from soil ranged from 10^{-3} to 10^{-5} while CRs reflecting U uptake from water containing elevated U concentrations ranged from 1 to 100, with the highest CR values being observed in leafy and root vegetables. In this experiment, U concentration in vegetation is reported based on dry-weight while the CR values are reported based on fresh-weight. Both of these studies confirmed the trend of a larger CR value for water/plant transfer rather than soil/plant transfer.

2.4 HEALTH EFFECTS

Natural U is classified as both a radiological and toxicological agent and is the only radionuclide for which toxicity is the limiting factor (Wrenn et al., 1985). U has been rated as "highly toxic" based upon the following definition (Goldwater, 1957): "Toxicity is the ability of a chemical molecule or compound to produce injury once it reaches a susceptible site in or on the body."

The establishment of a toxicological limit of U in the kidney has been challenging. A no-damage threshold of 3 μg U g^{-1} kidney has been generally accepted through the 1970s based upon studies conducted in the 1950s (Leggett, 1989). However, more current studies indicate that this threshold may be too high and there are numerous recommendations on establishing a new limit. In 1985, Wrenn et al. recommended using a threshold of 1 μg U g^{-1} kidney and incorporated a safety factor of 50 for a limit of 0.02 μg U g^{-1}. Kocher (1989) recommended using the 1 μg U g^{-1} threshold and incorporated a safety factor of 10 for a limit of 0.1 μg U g^{-1}. Legget (1989) provides an excellent review of this issue and recommended lowering the threshold of 3 μg U g^{-1} by a factor of 10 for a limit of 0.3 μg U g^{-1}. Finally, the International Commission on

Radiological Protection (ICRP) recommends a threshold of 1 μg U g^{-1} without a safety factor for a limit of 1 μg U g^{-1} (ICRP, 1992).

U presents a very low radiological risk due to a low specific activity, and its biological action is considered to be primarily a stable element (Wrenn et al., 1985). The ICRP 60 (1995) and NCRP 91 (1987) recommend an annual effective dose equivalent to the public of 100 mrem yr^{-1} for chronic exposures from all sources. In 1986, the Nuclear Regulatory Commission recommended that this limit be raised to 500 mrem yr^{-1} (Kocher, 1989).

Perhaps the most important factor influencing the potential health impact of natural U is its solubility. Ingestion absorption fractions and dose conversion factors have been established for three different solubility classifications (d-daily clearance, w-weekly clearance, and y-yearly clearance) while inhalation absorption fractions are classified in terms of clearance rates as fast, moderate, or slow. Solubility affects both the availability and exposure pathways of U with the route of exposure determining the health impact. Soluble U predominantly follows the ingestion pathway while the insoluble form tends to follow the inhalation pathway. Soluble U follows the ingestion pathway where it enters the blood stream, via the gastrointestinal (GI) tract, and freely moves throughout the body, while insoluble U is usually inhaled and is primarily retained in the lung compartment (Stannard, 1988).

2.4.1 The Ingestion Pathway

The absorption of U into a biological system is dependent on a number of factors such as nutritional state, intestinal content, age (Bosshard et al., 1992), and the level of intake (Wrenn et al., 1985). According to Wrenn et al. (1985), maximum absorption occurs at lower levels of intake rather than at higher levels of intake. Internally, ingested U faces three possible fates: rapid elimination, absorption with short-term retention, and adsorption with long-term retention. The majority of the ingested quantity, approximately 95%, is cleared via renal excretion with a biological half-life of 2 to 6 days (Bosshard et al., 1992). Based on numerous studies involving both animal and human subjects, it is currently thought that roughly 2% of ingested soluble U is absorbed into the GI tract (ICRP 78, 1997) although it may be as low as 1% (Durbin, 1998).

Absorbed U is available to all tissues in the body via blood transport. The target tissues of U, in terms of potential biological damage, are bone and kidney. It is in these

two organs that the two actions of U, radiological and toxicological, are observed.

2.4.2 Uranium in Bone

Due to long-term retention and half-life of U, the risk to bone is radiological (Bosshard et al., 1992). U is initially deposited on bone surfaces, especially growing surfaces (Durbin, 1998). Under equilibrium conditions, as is the case with chronic exposures, U is widely distributed through the bone volume (Wrenn et al., 1987). This redistribution is the result of U being buried by the growth of new bone surface, which allows for slow diffusion through the bone volume (Durbin, 1998). Although U is a known volume seeker, there is no evidence directly relating U to cancer induction in humans (Mayes and Rowland, 1985; Wrenn et al., 1985; Wrenn et al., 1987; Stannard, 1988; Bosshard et al., 1992).

ICRP 30 (1979) recommends the use of a two-compartment model based on calcium to describe U retention due to the similarity in skeletal kinetics thus allowing use of the general metabolic model for alkaline earth elements. In this model, 20% of the absorbed fraction is deposited in bone with a biological half-life of 20 days and 2.3% of the absorbed fraction was deposited in bone with a biological half-life of 5000 days. In a more recent publication (ICRP 69,

1995) "it is assumed that 15% of U leaving the circulation deposits on bone surfaces" with a biological half-life of 5 days. Approximately half of the U leaving the bone surface is returned to plasma while the other half moves to the "exchangeable bone volume (EBV)" compartment, which has a biological half-life of 30 days. Roughly 75% of the U leaving the EBV compartment is returned to the bone surface while the remaining 25% is committed to the "non-exchangeable bone" compartment with a biological half-life of 5000 days (ICRP 69, 1995).

2.4.3 Uranium in the Kidney

The obvious result of having a 2% absorption fraction is that approximately 98% of ingested U is passed through the kidneys and eliminated from the body. Additionally, the kidneys continue to filter the amount remaining in the blood, thus effectively filtering nearly 100% of ingested U.

A two-compartment model, similar to the bone model, describes deposition and retention of U in the kidney (ICRP 30, 1979). In the ICRP 30 model, 12% of the absorbed U is deposited and retained with a 6-day half-life, and 0.00052% is deposited and retained with a 1500 day half-life. According to ICRP 69, 63% U in the bladder contents results directly from circulation while an additional

12% enters from temporary deposition in the renal tubules. The 12% temporarily residing in the renal tubules is cleared with a 7-day half-life. This model accounts for 0.5% deposition in "other kidney tissue" with a half-life of 5 years, or 1825 days.

The importance of deposition and retention of U in the kidney is based on potential impairment of kidney function. The nephrotoxic action of U is a complex process and will not be discussed in detail here as it has been described by numerous authors (Voegtlin and Hodge, 1949; Tannenbaum, 1951; Leggett, 1989; Bosshard et al., 1992; Durbin, 1998). In general, free UO^{2++} can preferentially occupy Ca^{2+} binding sites in the brush-border membrane of the proximal tube where it can be incorporated into brush-border cells during the membrane renewal process (Durbin, 1998). Primary renal damage results in chemical changes in the blood and urine while secondary changes occur in structure or function of other tissues (Durbin, 1998). Extensive structural changes have been observed in the brush-border membrane after high dosages of U (Leggett, 1989). Damage to other portions of the kidneys may occur if the dosage is high enough, but, unless the damage is "severe," the kidneys normally recover completely (Stannard, 1988). Recovery from high doses of U have been found to result in the development of a tolerance to subsequent exposures (Leggett, 1989).

2.4.4 The Inhalation Pathway

The inhalation pathway is the primary route of exposure to the insoluble forms of radionuclides (Stannard, 1988), although it must be remembered that soluble forms are available through this pathway as well. Insoluble radionuclides tend to be immobile in biological systems due to their tendency to accumulate in soils where they are biologically available primarily through inhalation (Whicker and Schultz, 1986). U is known to accumulate in top soil and silt or clay fractions as a result of tight bonding with organic matter (Kirkham, 1979). Wrenn et al. (1985) report that U concentrations found in normal lungs are approximately 8 to 10 times higher than would be expected from human and animal injection studies. They concluded this is due to the inhalation of insoluble particles, which may be accountable for up to 85% of the U found in lungs.

In the case of natural U it is important to remember that, while there is no direct evidence linking it to cancer in humans, the inhalation pathway has produced malignant results in animal studies (Stannard, 1988) and cannot be discounted.

3. MATERIALS AND METHODS

3.1 STUDY PLAN

An experimental garden was established in the Nambe region, reproducing local growing conditions that could impact plant growth and U uptake. This involved developing an experimental design, establishing the garden, and determining the protocols for garden maintenance and produce collection. Experimental design involved selecting a study site and well water sources, collecting baseline soil, and choosing vegetable crop species. Establishing the garden required plot preparation, cage construction, and setting up the pots. Protocols for labeling and sampling water, soil, and vegetable crop samples were established before the study was initiated. Samples were submitted to the Soil and Water Testing (SWAT) Laboratory at New Mexico State University (NMSU) in Las Cruces, New Mexico. The SWAT analysis methods are referenced in Tables 2, 3, and 4.

3.2 EXPERIMENTAL DESIGN

The site was located at approximately longitude 36 and latitude 106 and central to the baseline soil collection sites and the experimental wells. It consisted of a small fenced field, located behind a private residence, which had not previously been irrigated or maintained. A padlocked gate was placed at the only entrance to the field and the homeowner, while allowing unlimited access to the researchers, helped control access by unauthorized persons.

Four wells were selected, including one control well (<1 µg U L^{-1}) and three treatment wells based upon location and U concentration. The control well was located in Los Alamos, NM, while the experimental wells were located in Nambe within a 0.5-km radius of the study site. The experimental wells, having concentrations of 150, 500, and 1200 µg U L^{-1} (Table 5), encompassed the range of U concentrations found by NMED (McQuillan, personal communication) in the Nambe region.

Approximately 50 18-L (5-gallon) baseline soil samples were collected from various locations in the region of interest that had not been previously irrigated. This soil was thoroughly mixed to form a composite baseline soil from which four samples were collected and analyzed for chemical (N, P, K, Ca, Mg, Na, pH, cation exchange capacity [CEC], and organic matter [OM]) and physical properties (% sand, silt, and clay) at the NMSU SWAT laboratory (Table 6).

Table 2. New Mexico State University Methods for Water Sample Analysis

WATER PARAMETERS	METHODS FOR WATER
Calcium	ICP
pH	EPA 150.1
Electrical Conductivity	EPA 120.1
Sodium	ICP
Magnesium	ICP
Potassium	ICP
Total Phosphorous	EPA 365.2
N-Nitrate	EPA 353.2
N-Nitrite	EPA 354.1
Ammonium as N	EPA 350.1
Kjeldahl N	EPA 351.2
Carbonate	EPA 310.1
Bicarbonate	EPA 310.1
Total Dissolved Solids	EPA 2540C
Total U	ICP-MS

Table 3. New Mexico State University Methods for Soil Sample Analysis

SOIL PARAMETERS	METHOD REFERENCES
Calcium	2
pH	6
Electrical Conductivity	2
Sodium	2
Magnesium	2
Potassium	1
Total Phosphorous	3 & 7
N-Nitrate	4
Ammonium	5
Kjeldahl N	5
Total U	4
Texture	Hydrometer
Cation Exchange Capacity	5

1. Chicek, L.J., Interpreting Soil Analysis, CES GUIDE A-126.
2. Diagnosis and Improvement of Saline and Alkaline Soils, Ed. L.A. Richards, USDA Handbook 60, February 1954.
3. Guide to Fertilizer Recommendations in Colorado, A.E. Ludwick and J.O. Reuss, Dept. of Agronomy, CSU, Fort Collins, Colorado, 1974.
4. Methods for Chemical Analysis of Water and Wastes, EPA 200.7, National Environmental Research Center, Cincinnati, Ohio, 1979.
5. Methods of Soil Analysis, Part 2, Chemical and Microbiological Properties, Ed., C.A. Black, ASA Monograph 9, Madison, Wisconsin, 1965.
6. Olsen, S.R., C.V. Cole, F.S. Watanabe, and L.A. Dean, Estimation of Available Phosphorous in Soils by Extraction with Sodium Bicarbonate, Circular No. 939, USDA, Washington D.C., March 1954.
7. Soil Survey, Laboratory Methods and Procedures for Collecting Soil Samples, SCS, USDA, 1972.

Table 4. New Mexico State University Soil and Water Testing Laboratory Methods for Vegetation Sample Analysis	
Parameter Analyzed	Test Methods
Standard Package	ICP
N-Nitrate	
Kjeldahl N	Block Digestion
Total U	ICP-MS

Tomato (*Lycopersicon esculentum*), squash (*Cucurbita pepo*), lettuce (*Lactuca scarriola*), and radish (*Raphanus sativus*) were selected based on common usage in home gardens and the USEPA's produce classification system. Selecting produce common to home gardens in the Nambe area provided data on U uptake, based on regional parameters, necessary for realistic risk estimations. Using USEPA classifications for garden produce provided a means of estimating annual ingestion rates and comparing U uptake based on defined classifications. This classification system categorizes produce into exposed, protected, or root crops (USEPA, 1995). Exposed vegetables are those grown above ground (lettuce and tomato), protected are those having a protective covering typically removed before consumption (squash), and root crops are those growing underground (radish).

3.3 GARDEN SETUP

A 7.3-m by 1.8-m (24-ft by 6-ft) chain-link cage with two padlocked doors in both 1.8-m (6-ft) sides was constructed to control human and animal access to the garden. The cage was positioned lengthwise from east to west to allow equal light distribution between treatment blocks. A low platform was built in the center of the cage providing 30- to 35-cm (12- to 14-in.) walkways lengthwise down each side of the cage. This elevated the pots off the ground to eliminate cross contamination from water drainage during treatment application.

Lattice was secured to the exterior walls and roof of the cage in order to provide partial shade for seedling plants. The cage was covered with clear four-mil plastic having roll-down flaps, which were lowered only during inclement weather, to control for cross contamination resulting from rain splash.

Sixty-four pots were filled with approximately 15 L (4 gallons) of baseline soil, labeled, and placed in the cage into four

Table 5. Chemical Properties of Nambe Well Water Containing Various Concentrations of Natural Uranium

Natural U Level (μg L^{-1})	Ca (mg L^{-1})	Mg (mg L^{-1})	Na (mg L^{-1})	K (mg L^{-1})	EC (dS m^{-1})	pH
<1	20	6.5	18	3.3	0.238	7.83
150	134	10.2	44	5.2	0.975	7.51
500	55	5.8	138	6.5	0.924	8.03
1200	55	5.8	199	9.6	1.220	7.93

Natural U Level (μg L^{-1})	NO$_3$-N (mg L^{-1})	NH$_4$-N (mg L^{-1})	TKN (mg L^{-1})	Carbonate (meq L^{-1})	Bi-Carbonate (meq L^{-1})	TDS (mg L^{-1})
<1	0.31	0.06	0.1	0.0	2.0	176
150	6.66	0.02	0.3	0.0	7.3	536
500	0.26	0.23	0.8	0.0	7.6	540
1200	0.41	0.00	0.9	0.0	5.2	712

Table 6. Chemical and Physical Properties of Baseline Soil and Soil Irrigated with Water Containing Various Concentrations of Natural Uranium

Treatment U Level (μg L^{-1})	Ca (mg Kg^{-1})	Mg (mg Kg^{-1})	Na (mg Kg^{-1})	K (mg Kg^{-1})	EC (dS m^{-1})	CEC (cmol (+) Kg^{-1})	pH
Baseline[a]	157 ± 16	16 ± 2.4	52 ± 4.6	118 ± 3.3	1.4 ± 0.1	8.2 ± 1.0	7.8 ± 0.1
<1	146	17	78[b]	94	1.3[b]	19	7.6
150	171	18	143	90	1.6	21	7.7
500	129	13	258	84	1.8	19	7.9
1200	245	30	437	104	3.1	19	7.9

Treatment U Level (μg L^{-1})	P (mg Kg^{-1})	NO$_3$-N (mg Kg^{-1})	NH$_4$-N (mg Kg^{-1})	TKN (mg Kg^{-1})	OM (g Kg^{-1})	U (mg Kg^{-1})	Texture
Baseline	20 ± 1.3	29 ± 3.3	7.3 ± 0.8	1680 ± 755	16 ± 2.5	2.3 ± 3.0	Sandy loam
<1	15	31	3.7	989	14[b]	1.6[b]	
150	15	22	3.6	946	15	1.9	
500	16	27	3.2	989	16	3.2	
1200	17	34	3.8	892	17	4.1	

[a]Baseline data includes $\pm$ SD.

[b]Denotes a significantly increasing trend with increasing U levels at the 0.05 probability level using the nonparametric Mann-Kendall Trend Analysis Test.

complete randomized blocks. The four blocks were separated by approximately 35 to 40 cm (14 to 16 in.) and were approximately 20 cm (8 in.) from the exterior cage walls. A thin layer of pea gravel, thoroughly rinsed with control water, was added on June 12 (approximately 3 weeks into the experiment) as an additional control for potential rain splash cross contamination.

The vegetable crops were planted in the 0.015 cubic meter (4-gallon) pots containing baseline soil on May 27, 1997. The plants were randomly assigned a position within each block by utilizing a random number table (Samuels, 1989). Each block had one of each species per well water treatment resulting in a block sample size of 16 (four plant species $\times$ four U concentrations) for a total experimental sample size of 64 (16 samples $\times$ 4 blocks).

Plants were watered as needed over the course of the experiment to field capacity. Watering occurred every other day for all species until July 19, at which time it was observed that field capacity was not being reached due to preferential flow to the sides of the pots. Appropriate steps were taken to correct this, which resulted in weekly watering for the remainder of the experiment.

Water was applied manually with a 4.8-L (1-gal.) color-coded container filled from the 49-L (13-gal.) Nalgene storage containers. This allowed controlled treatment application to prevent cross contamination through splash.

3.4 MAINTENANCE PROTOCOL

The first digit of NMED results for U concentration in the selected treatment wells was used to indicate water U concentration, although later testing by SWAT yielded different results. NMED U concentration results for the selected wells were 150, 350, and 900 μg U L^{-1}, which resulted in the following labels: (1) for 150 μg U L^{-1}; (3) for 500 μg U L^{-1}; (9) for 1200 μg U L^{-1}. The control well (<1 μg U L^{-1}) was indicated by (0). These labels (0, 1, 3, 9) were used to indicate the treatment level that the baseline soil and vegetable crops were exposed to.

A three-digit code was used to indicate pot location (block number 1 through 4), U concentration of water applied (0, 1, 3, 9), and vegetable crop species (1-radish, 2-lettuce, 3-squash, 4-tomato). For example, squash in block 4 irrigated with water containing 150 μg U L^{-1} would be labeled as 413 (block, water U concentration, and species type). Pots were also color-coded according to treatment water U concentration; control pots were unmarked (black), 150 μg U L^{-1} pots were marked

green, 500 μg U L^{-1} pots were marked yellow, and 1200 μg U L^{-1} pots were marked white.

Well water was analyzed for total U before and after the experiment while chemical (Ca, Na, Mg, K, P, nitrite/nitrate [NO_2-NO_3], ammonium [NH_4], total kjeldahl nitrogen [TKN], carbonates, bi-carbonates) and physical (pH, electrical conductivity [EC] and total dissolved solids [TDS]) properties were analyzed at the end of the experiment (Table 5). Samples were collected in labeled 500-mL polyethylene bottles, sealed with chain-of-custody tape, and sent to SWAT for analysis.

Control water was collected two times over the course of the experiment from a source located at LANL site Technical Area (TA) 21 in a 1100-L (300-gal) storage tank and transported to the garden site. Treatment waters were collected in numbered and color-coded 49-L Nalgene jugs over the course of the experiment and stored in a small locker at the garden site. Well water was collected as needed and stored for a maximum time of 12 days.

Light to all water storage containers was eliminated upon observed growth of red algae. Storage jugs were thoroughly rinsed with a spray nozzle, and the 1100-L water tank was thoroughly scrubbed and rinsed

during the second water collection to help reduce/eliminate algae growth.

Produce samples were collected at maturity using the produce sampling protocol established by the Ecology Group Soils and Foodstuffs Team (LANL, 1996). Produce was collected from lowest to highest irrigation treatment (i.e., control through 1200 μg U L^{-1}). Tomato and squash were collected on an individual basis while radish and lettuce crops were harvested at a single time. Tomatoes were also collected from two in-situ gardens being irrigated with water from the 150 and the 500 μg U L^{-1} treatment wells.

3.5 SAMPLE PROCESSING PROTOCOL

Soil samples collected before the experiment were analyzed for chemical (U, N, P, K, Ca, Mg, Na, pH, CEC, and OM) and physical (% sand, silt, and clay) properties. A 500-mL composite soil sample was collected for each treatment level by taking a grab sample from each pot based on its assigned treatment level. This resulted in a composite soil sample being submitted for the control, 150, 500, and 1200 μg U L^{-1} treatments. Samples were collected in labeled ziploc-type bags and sent to SWAT for analysis.

Samples collected at the end of the experiment were analyzed for the same

chemical properties analyzed before the experiment and for the depth distribution of U. The pots were allowed to freeze in a covered, secured outdoor location at TA-21 during September and October. Chilled pots, processed from the lowest to highest treatment level, were slit lengthwise, "peeled" open, and separated into three lifts. The "a" lift represented soil depth 0 to 3 in., the "b" lift represented soil depth 4 to 7 in., and the "c" lift represented the soil remaining in the pot. Lifts were combined, based on treatment level (0, 1, 3, 9) and depth (a, b, or c), to form a composite from which a 500-g grab sample was collected and submitted to SWAT for analysis. An error was made at this stage by not measuring depth of the bottom layer. This requires the assumption that the c-lift has the equivalent depth of a-lift and b-lift. This is a reasonable observation based upon personal observations. Soil sample data were not collected based on species, which would have increased the validity and reliability of U uptake data and CR estimations.

Produce samples were thoroughly rinsed with tap water, and radishes were scrubbed to remove as much dirt as possible and then hand-dried. Tomatoes were partially quartered, squash were sliced into 2.5-cm slices, and the radish top-growth and tap-root

were removed before placing samples into appropriately labeled paper bags for oven drying. Lettuce samples were dried for a minimum of 48 hours at 75°C while squash, tomato, and radish were dried for a minimum of 5 days at 75°C. Dried samples were stored until the end of the experiment when all produce and top-growth samples had been collected and processed. Top-growth for radish, squash, and tomato (collected at the end of the experiment) was processed in the same manner as the produce: thoroughly rinsed with tap water, hand-dried, and oven-dried.

Samples were ground using a Wiley mill with mesh #40 (38-mm) from lowest to highest treatment level based on species (tomato, lettuce, squash, radish). The ground samples were placed into labeled polyethylene containers, sealed with chain-of-custody tape, and sent to the NMSU SWAT laboratory for analysis.

3.6 URANIUM PLATING EXPERIMENT

During the course of the experiment, questions were raised concerning the potential for U plating onto the Nalgene water storage containers. The need to address this issue resulted in a secondary experiment to determine the amount of U lost due to the plating process.

This experiment was designed to replicate the conditions in which the Nambe well waters were stored during the garden study. During the garden experiment, waters were stored in 49-L Nalgene jugs for no more than 12 days at one time. Thus, it was necessary to determine the amount of U lost, due to plating, over a 12-day period. The critical aspect of this experiment involved minimizing the time between sample collection and sample testing. Therefore, all samples were shipped overnight and an agreement with the NMSU SWAT laboratory allowed for testing upon receipt of the samples.

Approximately 38 to 45 L of each experimental water were collected in a new, appropriately labeled Nalgene jug. The first 500-mL sample was taken at this time and shipped overnight for testing. Four subsequent water samples were collected, from each of the three treatment waters, every other day for 12 days and shipped immediately for analysis to SWAT.

4. DATA ANALYSIS AND ESTIMATIONS

4.1 PLANT UPTAKE

Background CR (Eq. 4-1) and experimental CR (Eq. 4-2) were estimated based on dry weight for both edible and inedible plant parts.

4.1.1 Concentration Ratio Formulas

4-1:   Background CR:

$CR = U_{bkg\ veg}\ (\mu g\ U\ Kg^{-1})\ /\ U_{bkg\ soil}\ (\mu g\ U\ Kg^{-1})$

4-2:   Experimental CR ($U_{net\ veg}$):

$CR = U_{net\ veg}\ (\mu g\ U\ Kg^{-1})\ /\ U_{net\ soil}\ (\mu g\ U\ Kg^{-1})$

Where:

$U_{net\ veg}\ (\mu g\ U\ Kg^{-1}) = U_{observed\ veg}\ (\mu g\ U\ Kg^{-1}) - U_{bkg\ veg}\ (\mu g\ U\ Kg^{-1})$

$U_{observe\ veg}\ (\mu g\ U\ Kg^{-1}) = U$ in vegetation after treatment

$U_{bkg\ veg}\ (\mu g\ U\ Kg^{-1}) =$ background U in vegetation

and

$U_{net\ soil}\ (\mu g\ U\ Kg^{-1}) = U_{trt\ soil}\ (\mu g\ U\ Kg^{-1}) - U_{bkg\ soil}\ (\mu g\ U\ Kg^{-1})$

$U_{observe\ soil}\ (\mu g\ U\ Kg^{-1}) = U$ in soil after treatment

$U_{bkg\ soil}\ (\mu g\ U\ Kg^{-1}) =$ background U in soil

4.2 RADIOLOGICAL DOSE ESTIMATIONS

Dose estimations were initially performed based upon gender and age categories including adult, pregnant woman, child age 1 to 10 years, and child 11 to 19 years. These estimations indicated adult dose was much greater than any of the other categories, largely because of water and vegetation ingestion rates. Therefore, radiological dose was estimated based on the target tissues of kidney, bone, "other tissues" (describing the remaining tissues in the body), and effective dose.

Radiological dose was estimated for kidney, bone, "other tissues," and whole

body based on exposure via well water ingestion, ingestion of produce irrigated with well water, and soil inhalation. Dose estimations were made using the metabolic model recommended by the ICRP 30 (1979), which accounts for both short- and long-term retention of U in target tissues. It was assumed that the input variables, such as the annual ingestion rate, annual inhalation rate, and U concentration in water, produce, and soil, were constant for the duration of exposure. This assumption, although highly unlikely, was intended to produce "worst-case exposure" dose estimations. Absorption fractions and dose conversion factors (DCFs) relating to ingestion were obtained from ICRP 69 (1995) while those relating to inhalation were obtained from ICRP 72 (1996). Variable parameters (Table 7) relating to ingestion rates of water and produce and soil inhalation were obtained from the USEPA Exposure Factors Handbook (1997a, 1997b, and 1997c) and Fresquez et al. (1996).

The bone surface DCF, rather than red marrow DCF, was used to convert the amount of activity deposited in bone. This decision was based on literature review information stating that U is plated on the bone surface before being incorporated into the bone structure itself (Durbin, 1998). The weekly (W) clearance rate class was used for water and vegetation in order to obtain conservative results. The clearance rate class for soil inhalation depended on whether the mean or maximum dose was being estimated.

Table 7. Input Factors for Mean and Maximum Ingestion and Inhalation Rate Calculations

Source	Mean	Maximum
Water[a] (L yr^{-1})	515	858
Tomato[a,b] (Kg yr^{-1})	31.4	90
Lettuce[a,b] (Kg yr^{-1})	11.3	22
Squash[a,b] (Kg yr^{-1})	24.5	138
Radish[a,b] (Kg yr^{-1})	19.6	99.0
Mass Loading[c] (Kg m^{-3})	9×10^{-8}	9×10^{-8}
Inhalation Rate[a] (m^3 yr^{-1})	40.8	40.8
Respirable Fraction	1.0	1.0
Duration of Exposure[c] (yr)	0.007	0.210

[a]USEPA (1997a, b, c), [b]Fresh mass basis, [c]Fresquez et al. (1996).

Mean and maximum doses were estimated for each exposure pathway. Mean soil inhalation dose was based upon the mean time of exposure (USEPA, 1997b) and by using the fast/moderate clearance rate absorption fraction and conversion factor (ICRP 72). Maximum soil inhalation dose was estimated based upon the mean time of exposure plus two standard deviations and by using the slow clearance rate absorption fraction and conversion factor (ICRP 72). Mean and maximum water doses were estimated using ingestion factors recommended by the USEPA Handbook of Exposure Factors (1997a). Mean vegetation dose was based on average consumption rates for species (lettuce and tomato) or produce type (radish-root and squash-protected) and on the western region (USEPA, 1997c). It is assumed that the average consumption rates for radish and squash were grossly overestimated because they were based on produce type rather than species and therefore included the average annual ingestion of all root crops and all protected crops. Maximum vegetation doses were estimated based on average consumption rates plus two standard deviations rather than on the 90th percentile value provided by the USEPA (1997c). This also contributed to an overestimation of dose contributed by vegetable ingestion.

The annual ingestion rate (R_{ingest}) describes the sum of the annual ingestion rate of well water and the annual ingestion rate of produce irrigated with well water (Eq. 4-3).

4-3: Annual Rate of Ingestion

R_{ingest} (Bq) = f_1 * r * C

Where: f_1 = fraction absorbed to the GI tract (unitless)

r = ingestion rate (Kg yr^{-1}) or (L yr^{-1})

C = concentration (Bq Kg^{-1}) or (Bq L^{-1})

The annual rate of inhalation (Bq) was estimated using Eq. 4-4.

4-4: Annual Rate of Inhalation

$R_{inhalation}$ (Bq) = f_1* ML * C_{Soil} * IR *RF * T

Where: f_1 = fraction absorbed to the GI tract (unitless)

ML = mass loading in air (Kg/m^3)

C_{Soil} = soil U concentration (Bq Kg^{-1})

IR = inhalation rate (m^3 yr^{-1})

RF = respirable fraction (unitless)

T = time (yr)

The annual ingested and inhaled activities were summed to obtain a constant value representing the total annual activity absorbed by the GI tract from the ingestion and inhalation pathways. The short- and long-term absorbed fractions for bone, kidney, and "other tissues" were obtained by multiplying the fraction absorbed by the GI tract by the short- and long-term absorption

fractions for each target tissue. The short- and long-term absorbed fractions of bone, kidney, and "other tissue" were summed to obtain a short- and long-term absorbed fraction for the whole body.

Short- and long-term doses were estimated for each target organ using a simple rate equation (Eq. 4-5). The total annual activity absorbed by the target tissues was based on retention time and the target organs' effective half-life, which was dependent on the retention time of each tissue (Eq. 4-6). Initial dose was estimated for one year, while the committed effective dose equivalent (CEDE) was calculated for adults at 50 years and children at 70 years as defined by ICRP 72.

4-5: Dose (Bq)

$Q(t) = R_{ingest}/k_{eff} * (1-e^{(-keff*t)})$

Where: R_{ingest} = the rate of ingestion (Bq/yr)

k_{eff} = effective rate loss constant (yr-1)

t = time (yr)

4-6: Effective Half-life

$T_{eff} = (T_{bio} + T_{phys})/2$

Where: T_{bio} is the biological half-life (yr^{-1})

T_{phys} is the physical half-life (yr^{-1})

4.3 URANIUM CONCENTRATION IN THE KIDNEY

The U concentration in the kidney was obtained by converting the activity (Bq) estimated in Eq. 4-6 to grams uranium (g U) using the specific activity of U (1.24E4 g U Bq^{-1}) (Shleien, 1992). Grams U were converted to µg U and divided by the 310 g, the mass of Reference Mans kidney (Shleien, 1992), which resulted in toxicity (µg U g^{-1} kidney).

4.4 STATISTICS

With the small sample size (n = 4) it was difficult to determine if the data were parametric or non-parametric. Through the process of producing frequency histograms, it was found that the four data points could be manipulated in such a way, via bin size, to approximate a normal distribution depending upon the biases of the researcher. The decision was made to treat the data as non-parametric based on the assumption that, as ecological samples, with a larger sample size the data would most likely be non-parametrically distributed. It must be noted that with the small sample size the power of the statistical tests is limited.

4.4.1 Statistical Tests

The Wilcoxon Rank Sum test (Hollander and Wolfe, 1973) was used to identify differences, at the 0.05 probability level, between pre- and post-treatment soil parameters, vegetation parameters at different treatment levels, and U uptake within and between species.

The Kendall's Tau Coefficient test (Gibbons and Chakraborti, 1992) was used to measure the association and significance of the association between the chemical/physical parameters in soil and well water based on the U concentration in well water.

The Friedman's Method for Randomized Blocks (Sokal and Rohlf, 1969) was used to identify significant differences in the soil U concentration with respect to soil

The Mann-Kendall Trend Analysis test (Gilbert, 1987) was used to evaluate the U plating data for trends in total U concentration over time in well water.

4.4.2 Concentration Ratio Analysis

Statistical analysis of the edible CRs was limited to the estimation of the variance (Eq. 4-7), error (Eq. 4-8), and standard deviation.

4-7: Variance of a Ratio

$$V(R) = (^1/_n)*(R^2)*[(S^2_y/y^2 + S^2_x/x^2)] - [(2*r_{xy}*S_y*S_x)/(y * x)]$$

Where: $R = {^x/_y}$
N = sample number
S^2_y = the y sample variance
S^2_x = the x sample variance
$\overline{y}$ = the y sample mean
$\overline{X}$ = the x sample mean
S_y = the y sample standard deviation
S_x = the x sample standard deviation
r_{xy} = the correlation coefficient between the

x sample and the y sample

The standard deviation (S_R) was estimated by taking the square root of the value obtained in Eq. 4-7.

4-8: Standard Error

$$SE = {^{SR}/} \text{sqrt (n)}$$

5. RESULTS

5.1 TREATMENT WATER

The average U concentrations in treatment well waters, based on pre- and post-experiment analysis, were 0.98, 150, 520, and 1200 µg U L^{-1}. Post-experiment samples were analyzed for Ca, pH, EC, Na, Mg, K, P, NO_2-NO_3, NH_4, TKN, carbonates, bi-carbonates, and TDS (Table 5). Nambe well water had pH levels ranging between 7.5 and 8.0, values which correspond with the formation of uranyl di- or tri-carbonate species (De Vivo et al., 1984). These are the dominant species found in Nambe well water (McQuillan and Montes, 1998)

5.2 URANIUM PLATING EXPERIMENT

A significant decreasing trend (p = 0.05) was observed in U concentration in the 150 µg U L^{-1} treatment water (Table 8). A non-significant decreasing trend was observed in the 500 µg U L^{-1} well water, while no trend was observed in the 1200 µg U L^{-1} well water.

Table 8. Uranium Concentrations in Stored Treatment Waters

Sample No.	150 μg U L^{-1}	500 μg U L^{-1}	1200 μg U L^{-1}
17-Aug	134.3[a]	511.2	1374.9
20-Aug	133.2	518.0	1379.0
24-Aug	133.6	512.9	1383.7
26-Aug	130.0	507.8	1373.0

[a]Denotes a significantly decreasing trend at the 0.05 probability level using the nonparametric Mann-Kendall Trend Analysis Test.

5.3 SOIL

The average soil texture was consistent with sandy loam, and the average baseline U concentration was 2321 μg Kg^{-1} (Table 6). Two opposing trends were observed in the overall U concentration with respect to soil irrigated with control water versus soil irrigated with the three Nambe treatment waters. The overall U concentration decreased in soil irrigated with control water (<1 μg U L^{-1}) while it increased linearly with increasing U concentration in Nambe well water.

The Mann Kendall Trend Analysis test indicates six parameters show significant increasing trends between pre- and post-treatment soil parameters. Na, EC, OM, and U were significant at the $\alpha = 0.05$ level while pH and P were significant at the $\alpha = 0.20$ level.

5.3.1 Soil Depth Analysis

As with total U concentration in soil, the depth analysis indicated two opposing trends with respect to soil irrigated with control water and soil irrigated with Nambe well waters. Soil irrigated with control well water showed a significant increase ($0.02<p<0.01$) in U concentration with increasing soil depth while soil irrigated with the 500 and 1200 μg U L^{-1} well waters showed a significant decrease ($0.02<p<0.01$) in U concentration with increasing soil depth (Table 9). The same decreasing trend was observed in soil irrigated with 150 μg U L^{-1} at $0.05<p<0.02$.

5.4 PRODUCE AND VEGETATION ANALYSIS

The general order of uptake among the four experimental produce species was radish≥lettuce>squash>tomato. Uptake in tomato and squash were significantly different from each other and from lettuce

and radish while no significant differences (p<0.05) were found between radish and lettuce (Table 10). A regression analysis showed U uptake within species was highly correlated to U concentration in water. Uranium concentrations in inedible plant portions indicate a significant increasing trend at $\alpha = 0.05$ in tomato, squash, and radish (Table 11).

Six parameters in lettuce were highly significant with respect to treatment level. Phosphorous (0.02<p<0.01), sulfur (0.05<p<0.02), and sodium (0.10<p<0.05) increased with increasing U concentration in irrigation water while calcium (0.05<p<0.02), aluminum (0.05<p<0.02), and potassium (0.20<p<0.10) decreased with increasing U concentration in irrigation water.

Phosphorous (0.01<p<0.001), aluminum (0.02<p<0.01), and calcium (0.20<p<0.10) exhibited significantly decreasing trends in edible radish while sodium (0.02<p<0.01) and sulfur (0.20<p<0.10) exhibited significantly increasing trends. Sodium (0.10<p<0.05) was the only parameter to exhibit an increasing trend in tomato while potassium, boron, and iron exhibited decreasing trends (0.20<p<0.10). Only one parameter showed a slightly significant relationship with respect to treatment level in squash. Total kjeldahl nitrogen (0.2<p<0.10) exhibited an increasing relationship to increasing U concentration in irrigation water.

Mean CRs ranged between 10^{-2} to 10^{-4} for edible (Table 12) and inedible (Table 13) portions of the four produce species, falling within recommended IAEA (1994) values. Estimated experimental CRs in radish (1.4 to 1.6) and lettuce (1.1 to 1.4) irrigated with Nambe well water exceeded recommended IAEA (1994) values by a factor of 100 while tomato (10^{-2}) and squash (10^{-1}) remained within IAEA (1994) values. While CRs were higher in inedible (top-growth) portions in tomato (0.2 to 0.5) and squash (0.6 to 1.3) and slightly lower in radish (0.7 to 1.0), the differences were not significant (p<0.05). Observed differences in uptake between species were consistent with Morishima et al. (1977), who found leafy vegetables (lettuce) and root vegetables (radish) have higher U uptake than berries (tomato and pumpkin), and Lakshmanan and Venkateswarlu (1988) who found uptake in radish root was greater than uptake in bottle gourd.

Table 9. Uranium Concentration (μg U Kg^{-1}) in Soil Based on Depth

Block	<1 μg U L^{-1} 0 to 3"	<1 μg U L^{-1} 3" to 6"	<1 μg U L^{-1} 6" to remainder
1	1660	1758	1854
2	1853	1893	1832
3	1817	2475	2498
4	2353	2427	2535

Block	150 μg U L^{-1} 0 to 3"	150μg U L^{-1} 3" to 6"	150 μg U L^{-1} 6" to remainder
1	2673	2673	2458
2	2910	2458	2303
3	2837	2535	2388
4	2588	2606	2513

Block	500 μg U L^{-1} 0 to 3"	500 μg U L^{-1} 3" to 6"	500 μg U L^{-1} 6" to remainder
1	3987	2838	2637
2	3602	2675	2416
3	3657	3346	3335
4	3927	3477	2786

Block	1200 μg U L^{-1} 0 to 3"	1200 μg U L^{-1} 3" to 6"	1200 μg U L^{-1} 6" to remainder
1	5934	3546	2902
2	4969	3538	2822
3	5911	4331	3170
4	6701	4692	3709

Table 10. Mean (± SD) U Concentrations in Edible Crop Tissue Irrigated with Water Containing Various Levels of Natural U (µg U Kg^{-1} dry weight)

Water U Concentration (µg U L^{-1})	Tomato	Squash	Radish	Lettuce
<1	8 ± 2.8C[a]	13 ± 2B	82 ± 18A	79 ± 20A
150	18 ± 3.5C	45 ± 19B	495 ± 193A	441 ± 140A
500	38 ± 1.9C	161 ± 70B	1306 ± 392A	1370 ± 191A
1200	67 ± 22.0C	285 ± 49B	2879 ± 830A	2304 ± 393A

[a]Means within the same row followed by the same upper-case letter were not significantly different at the 0.05 probability level using a nonparametric Wilcoxan Rank Sum test.

Table 11. Uranium Concentrations in Inedible Crop Portions Irrigated with Water Containing Various Levels of Uranium (µg U Kg^{-1})

Water U Concentration (µg U L^{-1})	Tomato	Squash	Radish	Lettuce
<1	64.3[*]	142.0*	60.4*	NA
150	144.6	168.3	278.5	NA
500	414.3	1165.0	865.6	NA
1200	666.0	2294.0	2340.5	NA

*Denotes a significantly increasing trend with increasing U levels at the 0.05 probability level using the nonparametric Mann-Kendall Trend Analysis Test.

Table 12. Mean (± SD) Concentration Ratios for Edible Portions Irrigated with Well Water Containing Various Levels of Natural Uranium

Water U Concentration (μg U L^{-1})	Tomato	Squash	Radish	Lettuce
< 1	$3.5 \times 10^{-3} \pm 7.0 \times 10^{-4}$	$5.6 \times 10^{-3} \pm 2.8 \times 10^{-4}$	$3.6 \times 10^{-2} \pm 4.7 \times 10^{-3}$	$3.4 \times 10^{-2} \pm 5.0 \times 10^{-1}$
150	$4.0 \times 10^{-2} \pm 1.0 \times 10^{-2}$	$1.3 \times 10^{-1} \pm 5.5 \times 10^{-2}$	$1.6 \times 10^{-0} \pm 5.3 \times 10^{-1}$	$1.4 \times 10^{-0} \pm 4.4 \times 10^{-1}$
500	$3.3 \times 10^{-2} \pm 9.9 \times 10^{-5}$	$1.6 \times 10^{-1} \pm 7.1 \times 10^{-2}$	$1.4 \times 10^{-0} \pm 4.6 \times 10^{-1}$	$1.4 \times 10^{-0} \pm 3.0 \times 10^{-1}$
1200	$2.9 \times 10^{-2} \pm 8.2 \times 10^{-3}$	$1.3 \times 10^{-1} \pm 4.6 \times 10^{-2}$	$1.4 \times 10^{-0} \pm 4.3 \times 10^{-1}$	$1.1 \times 10^{-0} \pm 3.9 \times 10^{-1}$

Table 13. Mean (± SD) Concentration Ratios for Inedible Portions Irrigated with Well Water Containing Different Levels of Natural Uranium

Water U Concentration (μg U L^{-1})	Tomato	Squash	Radish	Lettuce
< 1	$2.8 \times 10^{-2} \pm 6.2 \times 10^{-3}$	$6.1 \times 10^{-2} \pm 3.7 \times 10^{-3}$	$2.6 \times 10^{-2} \pm 4.9 \times 10^{-3}$	NA
150	$5.6 \times 10^{-1} \pm 1.4 \times 10^{-2}$	$6.5 \times 10^{-1} \pm 5.6 \times 10^{-2}$	$1.1 \times 10^{-0} \pm 4.6 \times 10^{-1}$	NA
500	$3.7 \times 10^{-1} \pm 9.4 \times 10^{-3}$	$1.3 \times 10^{-0} \pm 6.2 \times 10^{-2}$	$7.2 \times 10^{-1} \pm 4.7 \times 10^{-1}$	NA
1200	$2.0 \times 10^{-1} \pm 1.1 \times 10^{-2}$	$6.4 \times 10^{-1} \pm 4.7 \times 10^{-2}$	$8.6 \times 10^{-1} \pm 4.2 \times 10^{-1}$	NA

5.5 RADIOLOGICAL DOSE AND U IN KIDNEY

The estimated CEDE for an adult (50-yr exposure) exposed to 1200 μg U L^{-1} under maximum exposure conditions is 0.16 mSv (Table 14). As expected, bone received the highest dose of the three target tissues in both mean (50-yr: 0.1 mSv) and maximum (50-yr: 0.16 mSv) exposure conditions. The current regulatory guideline with respect to radiological dose is 1 mSv yr^{-1} with a 50-year cumulative exposure guideline of 50 mSv. Thus, under "worst case" exposure conditions (1200 μg U L^{-1}) with maximum water and produce ingestion and maximum inhalation, the estimated dose in 50 years is roughly 300 times less than the regulatory guidelines.

Evaluating the U concentration in kidney is challenging due to the varied recommended limits. The estimated U concentration in kidney under maximum conditions to water containing 1200 μg U L^{-1} is 0.8 μg U g^{-1} kidney (Table 15), a value which exceeds the recommended limits of Wrenn et al., (1985) (0.02 μg U g^{-1} kidney), Legget (1989) (0.3 μg U g^{-1} kidney), and Kocher (1989) (0.1 μg U g^{-1} kidney). However, this value does not recommend the limit established by the IAEA (1992) of 1 μg U g^{-1} kidney. Without further study on the toxicological limit of U in the kidney, it is impossible to draw a definitive conclusion regarding potential harm from exposure to the Nambe well water pathway.

The results of this study indicate the primary route of exposure in the Nambe well water pathway is ingestion of well water (99%) with ingestion of produce contributing roughly ~1% to the overall dose and kidney concentration.

Table 14. Estimated Committed Effective Dose Equivalents (CEDE) (mSv) for Mean and Maximum Exposures to Well Water Containing Different Levels of Natural Uranium via Water and Produce Ingestion and Soil Inhalation

	Kidney		Bone		Other Tissues		CEDE	
	Mean	Maximum	Mean	Maximum	Mean	Maximum	Mean	Maximum
<1 µg U L^{-1}								
50 Year	5.20×10^{-7}	1.1×10^{-6}	1.1×10^{-7}	2.4×10^{-4}	2.0×10^{-7}	4.2×10^{-7}	1.1×10^{-4}	2.4×10^{-4}
70 Year	5.20×10^{-7}	1.1×10^{-6}	1.2×10^{-4}	2.6×10^{-4}	2.0×10^{-7}	4.2×10^{-7}	1.2×10^{-4}	2.6×10^{-4}
150 µg U L^{-1}								
50 Year	5.4×10^{-5}	9.2×10^{-5}	1.2×10^{-2}	2.0×10^{-2}	2.0×10^{-5}	3.6×10^{-5}	1.2×10^{-2}	2.0×10^{-2}
70 Year	5.4×10^{-5}	9.2×10^{-5}	1.2×10^{-2}	2.0×10^{-2}	2.0×10^{-5}	3.6×10^{-5}	1.2×10^{-2}	2.0×10^{-2}
500 µg U L^{-1}								
50 Year	1.9×10^{-4}	3.2×10^{-4}	4.2×10^{-2}	7.0×10^{-2}	7.4×10^{-5}	1.3×10^{-4}	4.2×10^{-2}	7.0×10^{-2}
70 Year	1.9×10^{-4}	3.2×10^{-4}	4.4×10^{-2}	7.4×10^{-2}	7.4×10^{-5}	1.3×10^{-4}	4.4×10^{-2}	7.4×10^{-2}
1200 µg U L^{-1}								
50 Year	4.4×10^{-4}	7.4×10^{-4}	9.6×10^{-2}	1.6×10^{-1}	1.7×10^{-5}	2.8×10^{-4}	9.6×10^{-2}	1.6×10^{-1}
70 Year	4.4×10^{-4}	7.4×10^{-4}	1.0×10^{-1}	1.7×10^{-1}	1.7×10^{-5}	2.8×10^{-4}	1.0×10^{-1}	1.7×10^{-1}

Table 15. Estimated U Concentration in Kidney (µg U/g kidney) for Mean and Maximum Exposures to Well Water Containing Differing Levels of Natural Uranium via Water and Produce Ingestion and Soil Inhalation

Duration of Exposure (yr)	<1 (µg U L^{-1})	150 (µg U L^{-1})	500 (µg U L^{-1})	1200 (µg U L^{-1})
Mean Exposure				
1	3.2×10^{-4}	3.2×10^{-2}	1.2×10^{-1}	2.6×10^{-1}
50 and 70	5.6×10^{-4}	5.8×10^{-2}	2.0×10^{-1}	4.8×10^{-1}
Maximum Exposure[†]				
1	6.8×10^{-4}	5.6×10^{-2}	2.0×10^{-1}	4.6×10^{-1}
50 and 70	1.2×10^{-3}	9.8×10^{-2}	3.4×10^{-1}	8.0×10^{-1}

REFERENCES

Aieta, E.M., J.E. Singley, A.R. Trussell, K.W. Thorbjarnarson, and M.J. McGuire, "Radionuclides in Drinking Water: An Overview," The 1987 AWWA Annual Conference and Exposition, Kansas City, Missouri, June 14–18, 1987, pp. 144–152.

Agency for Toxic Substances and Disease Registry, *Toxicological Profile for Uranium*, ATSDR TP-90-29 (1990).

Bathija, A., Environmental Protection Agency, personal communication, November 1998.

Bosshard, E., B. Zimmerli, and Ch. Schlatter, "Uranium in the Diet: Risk Assessment of its Nephro- and Radiotoxicity," *Chemosphere* **24:3**, 309–321 (1992).

Cannon, H.L., "The Effect of Uranium-Vanadium Deposits on the Vegetation of the Colorado Plateau," *Am. J. Sci*. **250**, 735–770 (1952). (As quoted from S.C. Sheppard and W.G. Evenden, *J. Environ. Radioactivity* **8**, 1988.)

Cothern, R.C., and W.L. Lappenbusch, "Occurrence of Uranium in Drinking Water in the U.S.," *Health Physics* **45:1**, 89–99 (1983).

Cowart, J.B., and W.C. Burnett, "The Distribution of Uranium and Thorium Decay-Series Radionuclides in the Environment-A Review," *J. Environ. Qual.* **23**, 651–662 (1994).

Dement'yev, V.S., and N.G. Syromyatnikov, "Conditions of Formation of a Sorption Barrier to the Migration of Uranium in an Oxidizing Environment," *Geokhimiya* **4**, 459–465 (1968). (As quoted from J.K. Osmond, *Handbook of Environmental Isotope Geochemistry* [Elsevier Scientific Publishing Company, 1980], Chap. 7.)

De Vivo, B., F. Ippolito, G. Capaldi, and P.R. Simpson, *Uranium Geochemistry, Mineralogy, Geology, Exploration and Resources* (Stephen Austin/Hertford, England, 1984), pp. 89–98.

Drobkov, A.A., Usp. Sovrem. Biol. 31. (1951). (As quoted from H.L. Cannon, *Am. J. Sci*. **250**, 735, 1952.)

Dunn, C.E., *Mineral Exploration: Biological Systems and Organic Matter*, (Prentice-Hall, Englewood Cliffs, New Jersey, 1986), pp. 133–149. (As quoted from J.J. Mortvedt, *J. Environ. Qual*. **23**, 643-650, 1994.)

Durbin, P.W., "Health Physic Applications of Uranium," Forty-third American Academy of Health Physics Conference, Minneapolis, Minnesota, July 12–16, 1998, Continuing Education Program.

Eisler, R., "Radiation Hazards to Fish, Wildlife, and Invertebrates: A Synoptic Review," National Biological Service Contaminant Hazard Review Report 29 (1994), p. 124.

Eisenbud, M., and T. Gesell, *Environmental Radioactivity From Natural, Industrial, and Military Sources*, 4th ed. (Academic Press, San Diego, 1997).

Environmental Surveillance Program, "Environmental Surveillance at Los Alamos during 1995," Los Alamos National Laboratory report LA-13210-ENV (1996).

Fresquez, P.R., et al. 1996

Gibbons, J.D., and S. Chakraborti, *Nonparametric Statistical Inference*, 3rd ed., (Marcel Dekker, Inc., New York, 1992).

Gilbert, R.O., *Statistical Methods for Environmental Pollution Monitoring* (Van Nostrand Reinhold, New York, 1987.)

Goldwater, L.J., *Dangerous Properties of Industrial Materials* (Reinhold Publ. Corp. 1957). (As quoted from IAEA Tech. Report Series No. 15, 1963.)

Hess, C.T., J. Michel, T.R. Horton, H.M. Prichard, and W.A. Coniglio, "The Occurrence of Radioactivity in Public Water Supplies in the United States," *Health Physics* **48:5**, 553–586 (1985).

Hollander, M., and D.A. Wolfe, *Nonparametric Statistical Methods* (John Wiley and Sons, New York, 1973).

Hostetler, P.B., and R.M. Garrels, "Transportation and Precipitation of Uranium and Vanadium at Low Temperature, with Special Reference to Sandstone-type Uranium Deposits, *Econ. Geol*. **57**, 137–167 (1962). (As quoted from J.K. Osmond, *Handbook of Environmental Isotope Geochemistry*, [Elsevier Scientific Publishing Company, 1980], Chap. 7.)

International Atomic Energy Agency (1992)

International Atomic Energy Agency, Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments, Technical Report No. 364 (1994).

International Commission of Radiological Protection, Limits for Intakes of Radionuclides by Workers, ICRP publication 30, *Annals of the ICRP*, Part I: 2:3/4, (1979), (Pergamon Press, Oxford, 1979). (As quoted from D.C. Kocher*, Health Physics* **57:1**, 1989.)

International Commission of Radiological Protection, 1990 Recommendations of the International Commission on Radiological Protection, ICRP publication 60, *Annals of the ICRP,* No. 1-3; (Pergamon Press, New York, 1995). (As quoted from Shleien*, The Health Physics and Radiological Health Handbook*, 1992.)

International Commission of Radiological Protection, Age-dependent Doses to Members of the Public from Intake of Radionuclides: Part 3 Ingestion Dose Coefficients. ICRP publ. 69. *Annals of the ICRP* (Pergamon Press, New York, 1995).

International Commission of Radiological Protection, Age-dependent Doses to Members of the Public from Intake of Radionuclides: Part 5 Compilation of Ingestion and Inhalation Dose Coefficients. ICRP publ. 72. (Pergamon Press, New York, 1996).

International Commission of Radiological Protection, Individual Monitoring for Internal Exposure of Workers. ICRP publ. 78. (Pergamon Press, New York, 1997).

International Commission of Radiological Protection, Part I: Limits for Intakes of Radionuclides by Workers. ICRP publ. 30. (Pergamon Press, New York, 1979). (As quoted from Wrenn et al., Health Physics **48:5**, 1985.)

Irwin, R.J., M. Van Mouwerik, L. Stevens, M.D. Seese, and W. Basham, Environmental Contaminants Encyclopedia (1997). National Park Service, Water Resources Division, Fort Collins, Colorado. Distributed within the Federal Government as an Electronic Document (Projected public availability on the internet on NTIS: 1998.)

Kennedy, W.E., and D.L Strenge, Residual Radioactive Contamination from Decommissioning: Technical Basis for Translating Contamination Levels to Annual Total Effective Dose Equivalent, NUREG/CR-5512 (1992).

Kirkham, M.B., "Trace Elements" in *The Encyclopedia of Soil Science*, R.W. Fairbridge and C.W. Finkl, Eds. (Academic Press, Stroudsburg, Pennsylvania, 1979), Part I, Vol. 12, pp.571–575.

Kocher, D.C., "Relationship Between Kidney Burden and Radiation Dose from Chronic Ingestion of U: Implications for Radiation Standards for the Public," *Health Physics* **57:1**, 9–15 (1989).

Kotz, J.C., and K.F. Purcell, *Chemistry and Chemical Reactivity*, 2nd ed., (Saunders College Pub, Philadelphia, PA, 1991.)

Lakshmanan, A.R., and K.S. Venkateswarlu, "Uptake of Uranium by Vegetables and Rice," *Water, Air, and Soil Pollution* **38**, 151–155 (1988).

Leggett, R.W., "The Behavior and Chemical Toxicity of U in the Kidney: A Reassessment," *Health Physics* **57:3**, 365–383 (1989).

Linsalata, P., "Uranium and Thorium Decay Series Radionuclides in Human and Animal Foodchains-A Review," *J Environ. Qual.* **23**, 633–642 (1994).

Los Alamos National Laboratory, "Produce Sampling and Processing for the Foodstuffs Monitoring Program," Los Alamos National Laboratory Ecology Group hazard control plan, LANL-ESH-20-SF-HCP/OP-001 (1996).

Maassen, L.W., and S.L. Bolivar, "Uranium Hydrogeochemical and Stream Sediment Reconnaissance of the Albuquerque NTMS Quadrangle, New Mexico, Including Concentrations of Forty-Three Additional Elements," Los Alamos Scientific Laboratory report LA-7508-MS (1979).

Mahon, D.C., "Uptake and Translocation of Naturally-Occurring Radionuclides of the Uranium Series," *Bull. Environm. Contam. Toxicol.* **29**, 697–703 (1982).

Mayes, C.W., and R.E. Rowland, "Cancer Risk from the Lifetime Intake of Ra and U Isotopes," *Health Physics* **48:5**, 635–647(1985).

McGee, E.J., K.J. Johanson, M.J. Keatinge, H.J. Synnott, and P.A. Colgan, "An Evaluation of Ratio Systems in Radioecological Studies," *Health Physics* **70:2**, 215–221 (1996).

McQuillan, D., State of New Mexico Environment Department Ground Water Protection and Remediation Bureau, personal communication (March, 1996) (as quoted from Phil Fresquez).

McQuillan, D., and R. Montes, "Ground-Water Geochemistry Pojoaque Pueblo, New Mexico," New Mexico Environment Department Ground Water Quality Bureau, NMED/GWB-98/1 (1998).

Mengel, K., and E.A. Kirkby, *Principles of Plant Nutrition*, 2nd ed., (International Potash Institute, Worblaufen-Bern/Switzerland, 1979). (As quoted from S.C. Sheppard and W.G. Evenden, *J. Environ. Radioactivity* **8**, 255–285, 1988.)

Morishima, H., T. Koga, H. Kawai, Y. Honda, and K. Katsurayama, "Studies on the Movement and Distribution of Uranium in the Environments-Distribution of Uranium in Agricultural Products," *J. Radiat. Res*. **18**, 139–150 (1977).

Mortvedt, J.J., "Plant and Soil Relationships of Uranium and Thorium Decay Series Radionuclides-A Review," *J. Environ. Qual.* **23**, 643–650 (1994).

National Council on Radiation Protection and Measurements, "Recommendations on Limits for Exposure to Ionizing Radiation," NCRP Report No. 91 (1987). (As quoted from Kocher, *Health Physics* **57:1**, 1989.)

National Council on Radiation Protection and Measurements, "Exposure of the Population in the United States and Canada from Natural Background Radiation," NCRP Report No. 94 (1987).

Osburn, W.S., "Primordial Radionuclides: Their Distribution, Movement, and Possible Effect Within Terrestrial Ecosystems," *Health Physics* **11**, 1275–1295 (1965).

Osmond, J.K., *Handbook of Environmental Isotope Geochemistry* (Elsevier Scientific, Amsterdam, 1980).

Samuels, M.L., *Statisitics For The Life Sciences* (Dellen Publishing Company, San Francisco, 1989).

Sheppard, S.C., and W.G. Evenden, "Critical Compilation and Review of Plant/Soil Concentration Ratios for Uranium, Thorium, and Lead," *J. Environ. Radioactivity* **8**, 255–285 (1988).

Sheppard, M.I., and S.C. Sheppard, "The Plant Concentration Ratio Concept as Applied to Natural U," *Health Physics* **48:4**, 494–500 (1985).

Shleien, B., *The Health Physics and Radiological Health Handbook*, Revised Ed. (Scinta, Inc., Silver Spring, Maryland, 1992.)

Simon, S.L., and S.A. Ibrahim, *"*The Plant/Soil Concentration Ratio for Calcium, Radium, Lead, and Polonium: Evidence for Non-linearity with Reference to Substrate Concentration*," J. Environ. Radioactivity* **5**, 123–142 (1987). (As quoted from E.J. McGee et al. *Health Physics* 70:2. 1996.)

Sokal, R.R., and F.J. Rohlf, *Biometry* (W.H. Freeman and Company, San Francisco, 1969.)

Stannard, J.N., "Radioactivity and Health-A History," DOE/RL/01830-T59 (1988).

Tannenbaum, A., *"*Toxicology of Uranium," in *National Nuclear Energy Series*, div. IV, (McGraw-Hill, New York, 1951) Vol. 23; (As quoted from J.N. Stannard. Radioactivity and Health: A History, 1988. DOE/RL/01830-T59.)

Titayeva*,* N.A., *Nuclear Geochemistry* (Mir Publishers, Moscow, CRC Press, Boca Raton, 1994). pp. 51.

U.S. Environmental Protection Agency, 1995

U.S. Environmental Protection Agency, EPA Integrated Risk Information System (IRIS) electronic database. (Public domain information available from the government. EPA Criteria and Assessment Office, Cincinnati, OH, 1996a.)

U.S. Environmental Protection Agency-Office of Ground Water and Drinking Water, Radionuclides in Drinking Water (electronic publication, 1996b).

U.S. Environmental Protection Agency, Exposure Factors Handbook, EPA/600/P-95/002FB (1997a), Vol. I, Chap. 3.

U.S. Environmental Protection Agency, Exposure Factors Handbook, EPA/600/P-95/002FB (1997b), Vol. I, Chap. 5.

U.S. Environmental Protection Agency, Exposure Factors Handbook, EPA/600/P-95/002FB (1997c), Vol. II, Chap. 13.

Voegtlin, C., and H.C. Hodge, "Pharmacology and Toxicology of Uranium Compounds," in *National Nuclear Energy Series*, (McGraw-Hill. New York, 1949), div VI, vol.1, pts. III and IV. (As quoted from J.N. Stannard. Radioactivity and Health: A History, DOE/RL/01830-T59, 1988.)

Whicker, F.W., and V. Schultz, *Radioecology: Nuclear Energy and the Environment* (CRC Press, Boca Raton, Florida, 1986) Vol. I, pp. 132–133.

Wrenn, M.E., P.W. Durbin, B. Howard, J. Lipsztein, J. Rundo, E.T. Still, and D.I. Willis, "Metabolism of Ingested U and Ra," *Health Physics* **48:5**, 601–633 (1985).

Wrenn, M.E., P.W. Durbin, D.L. Willis, and N.P. Singh, "The Potential Toxicity of Uranium in Water," *J. AWWA*, 177–184 (1987).